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Determination of the ^{242}Pu Branching Ratio via Alpha-Gamma Coincidence

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Abstract

When the burn-up is high, the ^{242}Pu isotopic content becomes more important. The traditional correlation method [1] will fail. The ^{242}Pu isotopic content in the sample plays an essential role if the neutron coincidence method is used to quantify the total amount of plutonium. In one of the earlier measurements [2] we had a chance to measure an isotopic pure (> 99.95 %) ^{242}Pu thick sample and realized that the difference in the branching ratio (BR) value among current nuclear data[3] for the two important gamma-rays at 103.5-keV and 158.8-keV. In this study, the thick sample was counted on a 15% ORTEC safeguards type HPGe to further improve BR determination of the 159-keV gamma-ray. Furthermore, we have made a thin ^{242}Pu sample from the thick sample and performed alpha-gamma coincidence measurements. Our preliminary gamma-ray BR results are 4.37(6) E-4, 2.79(8) E-5, and 2.25(8) E-6 for 44.9-keV, 103.5-keV, and 158.9-keV, respectively.

Introduction

In the previous paper[2], we have mentioned that we were surprised to learn the difference in the branching ratio (BR) value among current nuclear data for the two important gamma-rays at 103.5-keV and 158.8-keV. For example, the Table of Radioactive Isotopes[3] values are 7.8(8) E-5 and 4.5(15) E-6, respectively; the IAEA evaluation[4] values are 2.63(9) E-5 and 3.0 (2) E-6, respectively. The IAEA evaluation results are predominately based on a study[5] by R. Vaninbroukx et al. The LLNL MGA code[1] uses the IAEA evaluated value for 103.5-keV and since the MGA code does not use 158.8- keV for ^{242}Pu determination, no information of the branching ratio of this peak is found in the code. A more recent publication[4] by Berlizov et. al. also reported that the BR of the 158.8-keV gamma-ray is lower by 35% at 2.20(8)E-6 via both gamma-ray measurement using a planar HPGe detector and a neutron coincidence measurement. Figure 1 shows a complied [8] decay scheme of ^{242}Pu. The alpha decay of ^{242}Pu populates states in ^{238}U at excitation energy of 0- keV, 44.91-keV, 148.41-keV, and 307.21-keV. The transition from 307-keV to 148-keV state produces gamma-ray with energy of 159-keV, etc. Because of small alpha branch to the higher excitation state and long half-life of ^{242}Pu (compared to the rest of plutonium isotopes) as well as cascade summing of the gamma-rays thus made the gamma-ray branching intensity difficult to determine. Significant differences of the three gamma-ray emission probability have been reported.
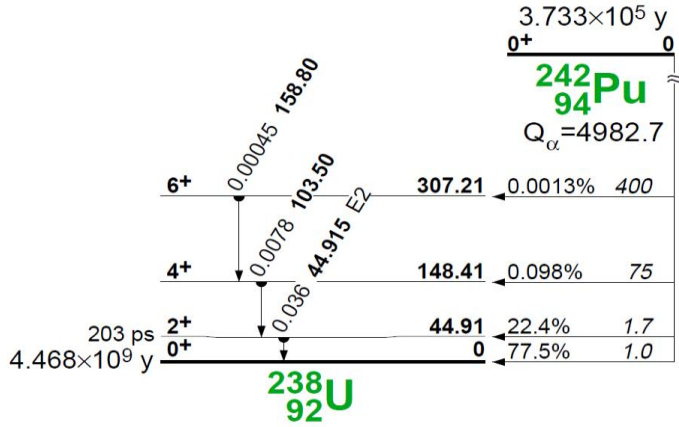
Figure 1. Alpha- and gamma-ray decay branch of ^{242}Pu taken from reference 8 compilation, most of the alpha decays to the first excited state (22.4%) and ground state (77.5%) in ^{238}U. Gamma-ray emission probabilities shown are per 100 decay of ^{242}Pu. The gamma-ray BR values are quite different from those in Ref 4.

Experimental setup

The ^{242}Pu sample was acquired from Oakridge National Laboratory with mass spectrometry results shown in Table 1 as well as decay corrected weight percentages to the present date, the t-half used in the calculations.

	Declared (08/01/07)	Present day (02/01/12)	t-half(yr)
Total PuO2 (4.53g)	Weight percentage		
238Pu	0.0032	0.0031	87.7
239Pu	0.0049	0.0049	24100
240Pu	0.0218	0.0218	6537.3
241Pu	0.0092	0.0074	14.35
242Pu	99.9589	99.9608	387000

This plutonium oxide sample (about 4.5 g in weight) was placed on the top of a ORTEC safeguards type HPGe detector (SGD-GEM-5030P4) that has energy resolution of ~ 650eV @ 120-keV and ~ 725 eV @ 160-keV. Digital data acquisition system, ORTEC DSPECpro, was used. A 30mil thick Cd absorber was placed between the sample and the HPGe detector to attenuate the 59-keV from the decay of ^{241}Am. Data were collected for 10-days. To minimize

effects due to summing and dead time (about 4% during the measurement), the sample was placed on the axis 15cm from the face of the detector.

A thin [242]Pu sample was also made by the same material evaporated on a thin platinum circular foil of 300mm^2 in area, 542 mg in weight, for particle-gamma measurement. The foil serves as the window to the vacuum system where an ORTEC silicon surface barrier (Si(SB)) detector of 150 μm thick 300mm^2 in area was mounted with its front facing the window. The evaporated side was facing the Si(SB) detector and the non-evaporated side was facing an ORTEC planar HPGe detector (GLP-36XXX) with energy resolution of 600 eV @ 120-keV. NIM electronics were used. Amplifiers, (ORTEC 572) with shaping time of 0.5 μs and 2 μs, were employed for Si(SB) and HPGe detectors, respectively. List-mode data acquisition system, FAST COMTEC MPA-3, was used for event-by-event recording with a time accuracy of 50 ns. List mode data were collected at for 15-days. The time and energy information was used to extract alpha-gamma coincidence information. This coincidence measurement was used to minimize [241]Pu (mostly beta-decay) gamma-ray at 44.20-keV, 44.86-keV and 103.68-keV contribution to the gamma-ray energy of interest at 44.92-keV and 103.50-keV. The alpha coincidence requirements also cleaned up the beta-related continuum in the planar HPGe detector. Two gamma-ray calibration standards, [109]Cd and [57]Co, were used to interpolate the detection efficiency at 103.5 keV. Figure 2 shows a spectrum of single gamma-ray measurement with the thick [242]Pu sample and Figure 3 showing a 2-D coincidence matrix with gamma-ray energy and particle energy measured with the thin [242]Pu sample.
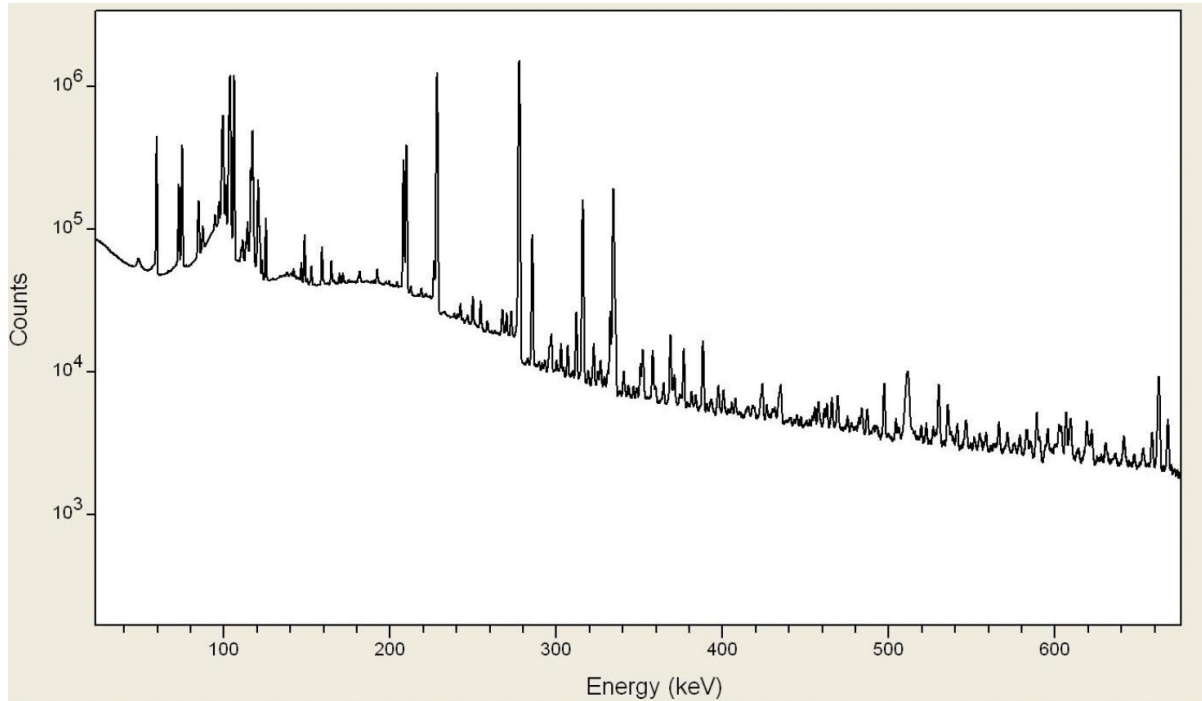

Figure 2. Gamma-ray spectrum of the thick [242]Pu sample, 30mil Cd absorber was used to attenuate 59-keV gamma-ray from the decay of [241]Am.
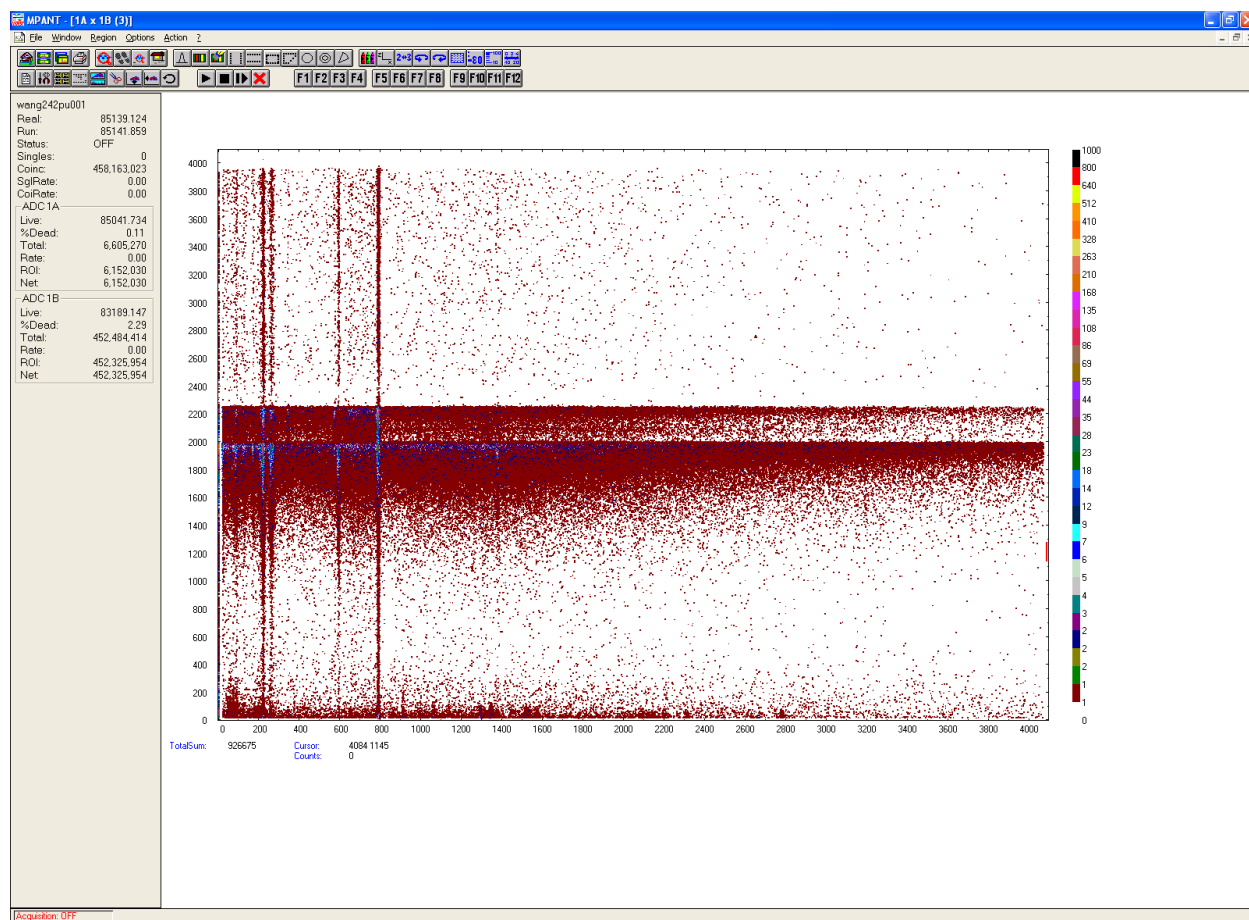
Figure 3. A 4096x4096 2D display of the particle gamma coincidence data, x-axis is gamma-ray energy and y axis is charged particle energy.

Data Analysis and results

a) **The 158.9-keV – Thick sample, 15% efficiency safeguards detector, via relative ratios**

A fit at the 158.8-keV region of the gamma-ray spectrum from the thick sample is shown in figure 4; the ^{242}Pu gamma-ray branching ratios were derived using the nearby ^{240}Pu and ^{241}Pu gamma-ray intensities with branching ratios from an LLNL evaluation and an IAEA evaluation. The results are shown in Table 2. The energy calibration of the spectrum was performed using a mixed gamma-ray standard and measured at 158.86 (3) keV. It is important to note that this BR is sensitive to the detector resolution, the purity of ^{242}Pu, the count rate and the tailing of the gamma-rays. Our preliminary result, 2.25(8) E-6, is in consistent with result from a recent measurement[6] of 2.20(8) E-6.
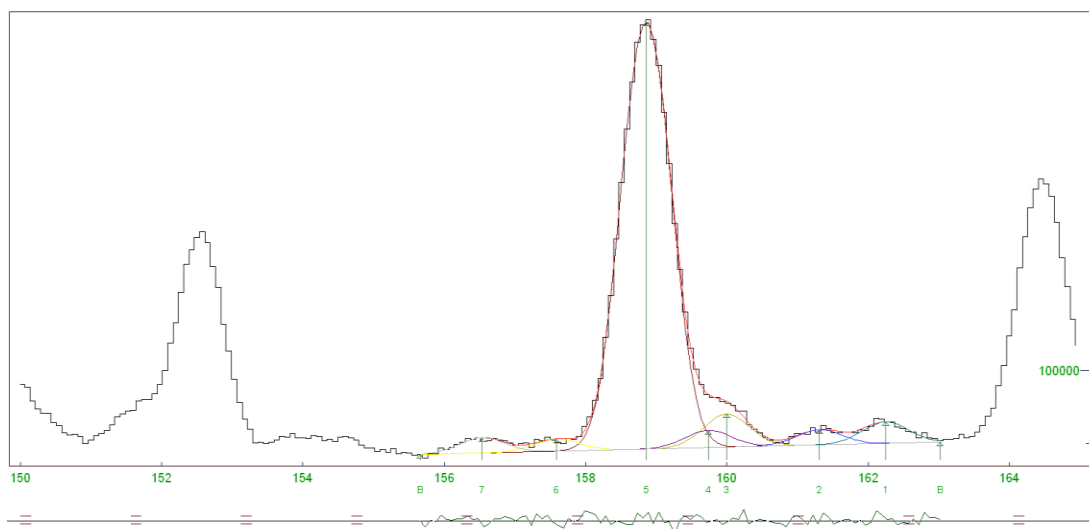
Figure 4. A fit to the 159-keV region from the thick ^{242}Pu sample measurement, the residuals are plotted at the bottom

	^{241}Pu BR	^{240}Pu BR	^{242}Pu(159) BR (derived)
LLNL(ref. 7)	6.74E-8	4.02E-6	2.25(8)E-6
IAEA (ref. 4)	6.58E-8	4.02E-6	2.29(8)E-6

Table 2. The ^{242}Pu 159-keV branching intensity results from this study

b) **The 44.9-keV – Thin sample, planar detector, alpha-gamma coincidence ratio determination with ^{238}Pu at 43.48-keV**

Because of close geometry of the counting and the beta-decay background, alpha particle coincidence data was used with an assumption of uniform alpha coincidence efficiency across the energy of interests. The alpha coincidence efficiency was determined to be 26.7(3) % by comparing the single and coincidence data. The BR ratios of the ^{238}Pu at 43.48-keV are quite similar between the IAEA evaluation and LLNL evaluation. The BR results of ^{242}Pu(44.9-keV) is shown in Table 3 and a fit to the coincidence data gated on alpha particles is shown in Figure 5. The energy of the ^{238}Pu and ^{242}Pu gamma-rays is 43.47(2) keV and 44.90(1) keV from the fit, respectively.

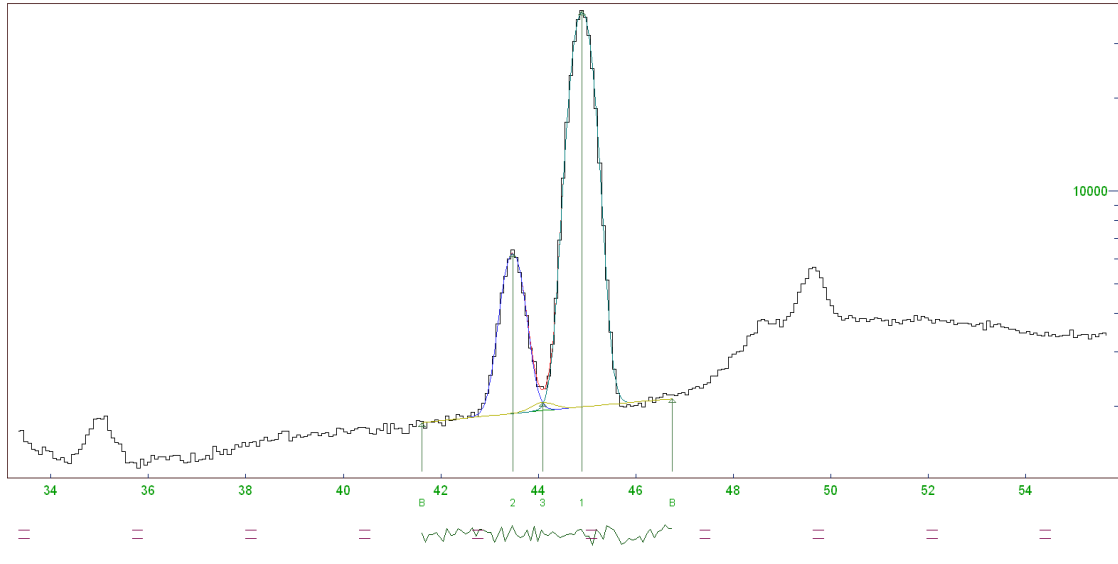Figure 5. The 45-keV region fit of the alpha-gamma coincidence gamma-ray data gated on the alpha particles

	^{238}Pu BR	^{242}Pu(44.9) BR (derived)
LLNL(ref. 7)	3.92E-4	4.37(6)E-4
IAEA (ref. 4)	3.94E-4	4.39(6)E-4

Table 3 BR of 44.9-keV gamma-ray derived from two different evaluations

c) **The 103.5-keV – Thin sample, planar detector, alpha-gamma coincidence, intra- , extra- polated efficiency determination via ratio determination with 46.5-keV**
Three gamma-ray calibration standards: ^{57}Co, ^{109}Cd, ^{241}Am, were used to interpolate the detector efficiency at 103.5-keV, and extrapolated to the 44.92-keV. The platinum foil is placed between these calibration standards and the detector. Using the branching ratio result from **b)** (LLNL) for 44.9-keV gamma-ray folding with proper efficiency corrections (1.75) and the fitting results of the alpha-gated gamma-ray areas, we have derived a BR of 2.79(8)E-5 for the 103.5-keV gamma-ray.

Conclusions

Using relative BR determination via known nearby gamma-ray decay from impurity isotopes (e.g. ^{241}Pu and ^{238}Pu, in this study) is strongly dependent on our knowledge of the BR these impurity isotopes. The results will vary depending on the reference BR used. The particle gamma-ray coincidence technique with list-mode data acquisition significantly improves the signal to background ratio for extracting area of the gamma-rays of interest. The gating on the alpha energy also reduces interferences of the beta-decay electrons and, especially, on our sample, the ^{243}Am interferences. We will improve the measurement by performing a chemical

separation of the ^{242}Pu sample in the future. Table 4 shows a compilation of BR results from various studies.

Energy(keV)	Table of Isotope [3]	IAEA [4]	Berlizov [6]	This study
44.9	3.6(5)E-4	3.72(7)E-4		4.37(6)E-4
103.5	7.8(8)E-5	2.63(9)E-5		2.79(8)E-5
158.9	4.5(15)E-6	3.0(2)E-6	2.20(8)E-6	2.25(8)E-6

Table 4 BR results from different studies.

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